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## DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] this invention relates to a thermally conductive pressure sensitive adhesive and the adhesion sheets which prepared this on the thermally conductive base material, and were made into gestalten, such as the shape of the shape of a sheet, or a tape.

[0002]

[Description of the Prior Art] electronic parts, such as a sealed type integrated circuit according from the former to a hybrid package, a multi-module, or a plastics metallurgy group, -- high integration of IC circuit etc. -- \*\* -- intermediary calorific value increases, and since there is a possibility that electronic parts may start a functional disorder for a temperature rise, the cure which attaches radiator material, such as a heat sink, to electronic parts, and prevents a functional disorder is taken

[0003] The method using the adhesives which added the aluminium powder etc. as a method of attaching radiator material to electronic parts to the constituent containing a polymerization nature acrylic-ester monomer and a free radical initiator is learned (U.S. Pat. No. 4,722,960 specification). However, for the above-mentioned adhesives, the problem that it is deficient in the manufacture efficiency of an electronic instrument that it is necessary to intercept oxygen and to carry out hardening processing, using a primer, and there is the need of carrying out temporary fixation of the adherend until it requires and hardens many times and many efforts to adhesion processing after painting this on both electronic parts, and radiator both [ one side or ] etc. is \*\*\*\*\*.

[0004] On the other hand, it is possible to attach radiator material to electronic parts simple, without also learning the method using the thermally conductive pressure sensitive adhesive equipped with thermal conductivity and the pressure-sensitive adhesive property, and adhesion processing taking many times and many efforts according to this. For this thermally conductive pressure sensitive adhesive, it is \*\* or intermediary \*\*\*\* that sufficient adhesive property is not acquired if high polarity monomers, such as an acrylic acid, are not used as a copolymerization monomer in order to secure the adhesive property to the material of a heat sink or a semiconductor package in which the most makes acrylic polymer adhesive polymer, and has the charge of facing of high polarity in this case.

[0005]

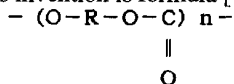
[Problem(s) to be Solved by the Invention] However, it is \*\*\*\*\* that viscosity elevation to the extent that coating cannot be carried out is seen, or an elastic modulus increases by elevation of cohesive force, and sufficient adhesive property cannot be secured by the polarity of the thermally conductive filler blended in the acrylic polymer using such a high polarity monomer in order to raise thermal conductivity. For this reason, the problem of the kind of thermally conductive filler which can be blended being restricted, or the loadings being restricted, and being unable to attain restrictions on manufacture and adhesive and thermally conductive highly efficient-ization is \*\*\*\*\*.

[0006] this invention aims at offering the adhesion sheets to the thermally conductive pressure sensitive adhesive which is excellent in the adhesive property and thermal conductivity which do not receive restrictions in the kind or loadings of a thermally conductive filler in the light of the above-mentioned conventional situation.

[0007]

[Means for Solving the Problem] By using the polymer which has polycarbonate structure as adhesive polymer, as a result of inquiring wholeheartedly, in order that this invention persons may attain the above-mentioned purpose Since sufficient adhesive property can be discovered and there are no restrictions in the kind and loadings of a thermally conductive filler in this case even if it does not use a high polarity monomer like acrylic polymer It is \*\*\*\*\* for being able to give desired thermal conductivity, knowing that the thermally conductive pressure sensitive adhesive which is excellent in both an adhesive property and thermal conductivity will be obtained after all, and completing this invention.

[0008] That is, this invention is formula [ to pour ]:

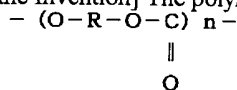


Polymer with the polycarbonate structure of having the repeat unit expressed with (R is the hydrocarbon group of the shape of a straight chain of carbon numbers 2-20, and the letter of branching), b) The thermally conductive pressure sensitive adhesive

characterized by including the thermally conductive filler of the rate which serves as the 10 - 300 weight section to this polymer 100 weight section (claims 1 and 2), The adhesion sheets (claim 3) which come to prepare the layer of the thermally conductive pressure sensitive adhesive of the above-mentioned composition in one side or both sides of a thermally conductive base material are started.

[0009]

[Embodiments of the Invention] The polymer of a component of this invention is following formula;



It is polymer with the polycarbonate structure of having the repeat unit expressed with (R is the hydrocarbon group of the shape of a straight chain of carbon numbers 2-20, and the letter of branching), and the polyester specifically compounded in the condensation reaction using a diol and/or a dicarboxylic acid with polycarbonate structure is mentioned.

[0010] There is polyester compounded by the condensation reaction with the polyester compounded by the condensation reaction with a diol with \*\* polycarbonate structure or its derivative, a dicarboxylic acid, or its derivative, a dicarboxylic acid with \*\* polycarbonate structure or its derivative, a diol, or its derivative among these polyester. Among these, especially the diol with polycarbonate structure has desirable polyester of \*\* of eye an available hatchet and the former as commercial elegance.

[0011] As a diol with the polycarbonate structure where it is used here, there are hexa MECHIRENKA-BONE-TOJIO-RU, 3-methyl pen TENKA-BONE-TOJIO-RU, a propylene carbonate diol, etc. those mixture or those copolymerization objects, etc. this diol has that good whose number average molecular weight is usually 800 (usually -- up to 10,000) or more preferably 500 or more as the commercial elegance of such a diol -- "PLACCEL CD208PL" by Daicel Chemical Industries, Ltd., "this CD210PL", "this CD220PL", "this CD208", "this CD210", "this CD220", and "-- this CD208HL" and "-- this CD210HL" and "-- this CD220HL" etc. can be mentioned

[0012] As a diol component, you may carry out mixed use of the components, such as a diol of the shape of a straight chain, such as ethylene glycol, a propylene glycol, butanediol, hexanediol, octanediol, the Deccan diol, and an OKUTA decane diol, and a diol of the various letters of branching, suitably as occasion demands. Moreover, in order to carry out macromolecule quantification of the polymer, you may carry out little addition of the polyol component of three or more organic functions.

[0013] The above-mentioned hydrocarbon group is a straight chain-like thing, a dicarboxylic-acid component is that to which the carbon number made the molecule skeleton the hydrocarbon group of the aliphatic series of 2-20, or an alicycle group, and its \*\*\*\*\* is [ \*\*\*\*\* is also the thing of the letter of branching and ] also good. Specifically, there are a succinic acid, a methyl succinic acid, an adipic acid, a PIMERITSUKU acid, an azelaic acid, a sebacic acid, 1, 12-dodecane diacid, 1, 14-tetradecane diacid, a tetrahydrophthal acid and methylene tetrahydrophthal acids, these acid anhydrides, low-grade alkyl ester, etc.

[0014] \*\* Polyester can be obtained by carrying out the condensation reaction (esterification reaction) of the above-mentioned diol component and the above-mentioned dicarboxylic-acid component using a non-catalyst or a proper catalyst according to a conventional method. although it is desirable to consider a diol component and a dicarboxylic-acid component as this mol reaction on the occasion of this reaction, in order to promote a reaction -- \*\* et al. -- \*\* -- or -- it may use superfluously and you may make it react

[0015] as molecular weight of the polymer of a components including such polyester, it is good that weight average molecular weight is 50,000 (usually -- up to 300,000) or more still more preferably 30,000 or more preferably 10,000 or more If molecular weight becomes small too much, since cohesive force is insufficient, if an adhesive property falls and it becomes large too much, neither which produces and burns a problem of the coating nature of a pressure sensitive adhesive is desirable.

[0016] As a thermally conductive filler of b component of this invention, ceramic powder, such as various metal powders, and an aluminum oxide, alumimium nitride, a titanium dioxide, a titanium boride, boron nitride, silicon nitride, silicon carbide, is various and usable. That is, by having used the specific polymer of the aforementioned a component as adhesive polymer in this invention, various things can be freely chosen as a thermally conductive filler, and when viscosity rises according to an interaction with a polymer polar group which was seen by acrylic polymer or an elastic modulus increases by elevation of cohesive force, there is especially no \*\*\*\*\* when.

[0017] Moreover, in a relation with polymer, there are also no loadings of the above-mentioned thermally conductive filler, and special restrictions can choose them in the latus range according to the thermal conductivity to need. Generally, it is good to make it the 10 - 300 weight section and become the 20 - 120 weight section preferably to the polymer 100 weight section of the above-mentioned a component. If good thermal conductivity is hard to be obtained and the 300 weight sections are exceeded, when this amount does not fulfill 10 weight sections, since an adhesive property etc. will be spoiled, it is not desirable.

[0018] In order that the thermally conductive pressure sensitive adhesive of this invention may contain the polymer of the above-mentioned a component, and the thermally conductive filler of b component at an above-mentioned rate and also may raise the maintenance property of a pressure sensitive adhesive, it can also construct a bridge using the proper bridge formation method. There is the method using the so-called cross linking agent of adding and carrying out crosslinking reaction of the compound which has the hydroxyl group and/or carboxyl group which are contained in the polymer of a components, such as the poly isocyanate system compound, an epoxy system compound, and an aziridine compound, and the basis which can react as a concrete means of the bridge formation method. Also in a cross linking agent, the poly isocyanate system compound is especially desirable.

[0019] As this poly isocyanate compound, ethylene di-isocyanate, Low-grade aliphatic poly isocyanates, such as butylene diisocyanate and hexamethylene di-isocyanate Cyclo pentylene diisocyanate, cyclohexylene diisocyanate, Alicycle group poly isocyanates, such as isophorone diisocyanate, 2, 4-tolylene diisocyanate, Aromatic poly isocyanates, such as diphenylmethane diisocyanate and xylylene diisocyanate, a tolylene diisocyanate addition product, a hexamethylene di-isocyanate addition product of a trimethylol propane, etc. are mentioned. These cross linking agents are independent, or can use one of them by two or more sorts of mixed stocks. It is good 0.5 - 5 weight section and that the amount used considers as 1 - 3 weight section preferably to the polymer 100 weight section of the aforementioned a component.

[0020] You may blend various kinds of well-known tackifiers besides the above-mentioned component with the thermally conductive pressure sensitive adhesive of this invention conventionally. By combination of a tackifier, there is a bird clapper that it is easy to balance a pressure-sensitive adhesive property and thermal resistance. Moreover, various kinds of conventionally well-known additives, such as the shape of fine particles, such as a common bulking agent, a pigment, and a flame retarder, and a particle and \*\*\*\*\*, can be arbitrarily included in the thermally conductive pressure sensitive adhesive of this invention as occasion demands. Furthermore, it is also possible to raise endurance by addition of various kinds of antioxidants.

[0021] The adhesion sheets of this invention prepare the above-mentioned thermally conductive pressure sensitive adhesive in one side or both sides of a thermally conductive base material, and it is taken as the gestalt of the shape of the shape of a sheet, or a tape. Here, thermally conductive plastic film, such as a sheet-like object of the polymer which is excellent in the thermal conductivity of metaled (an alloy is included) \*\*\*\*\* which is excellent in thermal conductivity, such as aluminum, copper, stainless steel, and a beryllium copper, thermally conductive silicon, etc. as a thermally conductive base material, polyester in which the thermally conductive filler was included, and a polyimide, is used.

[0022] Moreover, as a thermally conductive base material, the film of heat-resistant polymer, such as a polyimide, polyethylene terephthalate, polyethylenenaphthalate, a polytetrafluoroethylene, a polyether ketone, polyether sulphone, the poly methyl pentene, polyether imide, a polysulfone, polyphenylene sulfide, a polyamidoimide, a polyester imide, and an aromatic polyamide, can also be used as a heat-resistant film besides the above.

[0023] Although the thickness of a thermally conductive base material can be determined suitably, it is usually better than the point of heat-resistant [ in the state where the layer of a thermally conductive pressure sensitive adhesive was prepared ], and thermally conductive to be preferably referred to [ 10-125-micrometer ] as 25-100 micrometers. Moreover, although the layer thickness of the thermally conductive pressure sensitive adhesive prepared on this can also be determined suitably, it is usually better than points, such as an adhesive property and thermal conductivity, to be preferably referred to [ 10-200-micrometer ] as 30-130 micrometers. Of course, the layer thickness of a thermally conductive pressure sensitive adhesive is able for the thickness of a thermally conductive base material to exceed 125 micrometers, and to exceed 200 micrometers by the case.

[0024] What is necessary is just to perform manufacture of the adhesion sheets of this invention by imprinting this layer to one side or both sides of a thermally conductive base material, after applying a thermally conductive pressure sensitive adhesive and forming the layer of the thermally conductive pressure sensitive adhesive of request thickness on a separator. Moreover, the method of applying the above-mentioned thermally conductive pressure sensitive adhesive to one side or both sides of a thermally conductive base material directly, and forming the layer of the thermally conductive pressure sensitive adhesive of request thickness, without using a separator may be used. According to the kind of thermally conductive base material etc., a proper method is employable.

[0025] What is necessary is to infix the above-mentioned charge of a binder among both, and just to carry out sticking-by-pressure processing using the pressure-sensitive adhesive property, in order to carry out adhesion fixation of electronic parts and the radiator material using the thermally conductive pressure sensitive adhesive or its adhesion sheets of this invention. Thereby, adhesion fixation of electronic parts and the radiator material is carried out at thermally conductive fitness, the generating heat by electronic parts is efficiently transmitted to radiator material, and functional disorder-ization of the electronic parts resulting from the temperature rise is prevented.

[0026] There is a sealed type integrated circuit by IC chip, the hybrid package, the multi-module, the power transistor, and the plastics metallurgy group etc. in the electronic parts for adhesion fixation. In this invention, although IC circuit was accumulated highly, it is [ like ] applicable in favor of adhesion fixation of electronic parts with large calorific value. There are a heat sink which consists of metaled \*\*\*\*\*, sheet-like objects, etc., such as aluminum illustrated as formation material of a thermally conductive base material and copper, other radiators, etc. in the radiator material of another side. \*\*\*\*\* of a radiator is also good at the proper structures, such as a gestalt which has a cooling fin.

[0027] Business, such as the fixed purpose of the member in various fields, such as building materials, a car, an aircraft, and a vessel, can also be presented with the thermally conductive pressure sensitive adhesive or its adhesion sheets of this invention, and it can do so the same effect as the above also to these.

[0028]

[Example] Although the example of this invention is indicated and being explained more concretely below, the range of this invention does not receive a limit at all according to the following examples. In addition, that it is with the section means the weight section hereafter. Moreover, the weight average molecular weight of polymer is the measured value (polystyrene conversion) by the gel permeation chromatography.

[0029] It is 180 degrees C, attaching an agitator, a thermometer, and a water column to an example 1 4 TSU mouth separable flask, teaching this Djibouti rutin oxide (henceforth DBTO) 62mg as polycarbonate diol ["PLACCEL CD220PL" hydroxyl value:56.1 KOHmg/g] 250g, 26.1g of sebacic acids, and a catalyst, and stirring under existence of a small amount of toluene as a

reaction water ecchris solvent. [ by Daicel Chemical Industries, Ltd. ] The temperature up was carried out and it held at this temperature. After a while, outflow separation of water is accepted and a reaction began to avancé. The reaction was continued for about 25 hours and the polyester whose weight average molecular weight is 45,000 was obtained.

[0030] After diluting this polyester with toluene to 50 % of the weight of solid-content concentration, the hexamethylene di-isocyanate addition product ["coronate HL"] 2 made from Japanese Polyurethane section of a trimethylol propane was blended with the alumina (aluminum 2O3) 100 section as a cross linking agent per the solid-content (polyester) 100 section and as a thermally conductive filler, stirring mixture was improved, and the thermally conductive pressure sensitive adhesive was prepared.

[0031] This thermally conductive pressure sensitive adhesive was applied on the separator by the applicator, it dried for 5 minutes at 120 more degrees C after dryness for 5 minutes by 40 degrees C, and the layer of the thermally conductive pressure sensitive adhesive whose thickness is 50 micrometers was formed. This layer was imprinted to both sides of the aluminum foil whose thickness as a thermally conductive base material is 30 micrometers, and the adhesion sheet was produced.

[0032] The agitator, the thermometer, and the water column were attached to the example 2 4 TSU mouth separable flask, polycarbonate diol ["PLACCEL CD220PL" hydroxyl value:56.1 KOHmg/g] 500g, 50.6g of sebacic acids, and DBTO(catalyst)120mg were taught to this, the temperature up was carried out to 180 degrees C under existence of a small amount of toluene as a reaction water ecchris solvent, stirring, and it held at this temperature. [ by Daicel Chemical Industries, Ltd. ] After a while, outflow separation of water is accepted and a reaction began to advance. The reaction was continued for about 40 hours and the polyester whose weight average molecular weight is 48,000 was obtained.

[0033] After diluting this polyester with toluene to 50 % of the weight of solid-content concentration, the hexamethylene di-isocyanate addition product ["coronate HL"] 2 made from Japanese Polyurethane section of a trimethylol propane was blended with the titanium-boride (TiB2) 80 section as a cross linking agent per the solid-content (polyester) 100 section and as a thermally conductive filler, stirring mixture was improved, and the thermally conductive pressure sensitive adhesive was prepared. This thermally conductive pressure sensitive adhesive was used, and the adhesion sheet was produced like the example 1.

[0034] Except having changed the example 3 thermal-conductivity filler into the titanium-boride (TiB2) 100 section, like the example 1, the thermally conductive pressure sensitive adhesive was prepared, and the adhesion sheet was produced like the example 1 using this.

[0035] The agitator, the thermometer, and the water column were attached to the example 4 4 TSU mouth separable flask, polycarbonate diol ["PLACCEL CD210PL" hydroxyl value:115 KOHmg/g] 200g, 20.5g of succinic anhydrides, and DBTO(catalyst)100mg were taught to this, the temperature up was carried out to 180 degrees C under existence of a small amount of toluene as a reaction water ecchris solvent, stirring, and it held at this temperature. [ by Daicel Chemical Industries, Ltd. ] After a while, outflow separation of water is accepted and a reaction began to advance. The reaction was continued for about 27 hours and the polyester whose weight average molecular weight is 50,000 was obtained.

[0036] After diluting this polyester with toluene to 50 % of the weight of solid-content concentration, the hexamethylene di-isocyanate addition product ["coronate HL"] 2 made from Japanese Polyurethane section of a trimethylol propane was blended with the alumina (aluminum 2O3) 80 section as a cross linking agent per the solid-content (polyester) 100 section and as a thermally conductive filler, stirring mixture was improved, and the thermally conductive pressure sensitive adhesive was prepared. This thermally conductive pressure sensitive adhesive was used, and the adhesion sheet was produced like the example 1.

[0037] Except having changed the example 5 thermal-conductivity filler into the titanium-boride (TiB2) 80 section, like the example 4, the thermally conductive pressure sensitive adhesive was prepared, and the adhesion sheet was produced like the example 1 using this.

[0038] Except having changed the example 6 thermal-conductivity filler into the boron nitride (BN)80 section, like the example 4, the thermally conductive pressure sensitive adhesive was prepared, and the adhesion sheet was produced like the example 1 using this.

[0039] The monomer mixture of the example of comparison 1 butyl-acrylate 95 section and the acrylic-acid 5 section was used, the solution polymerization of the mixed solution which added the toluene 150 section and the azobisisobutyronitril 0.1 section into this monomer mixture was carried out at 60 degrees C among nitrogen atmosphere for about 7 hours, and the acrylic polymer solution whose solid-content concentration is 40 % of the weight was obtained.

[0040] In this acrylic polymer solution, the tolylene diisocyanate addition product ["coronate L"] 2 made from Japanese Polyurethane section of a trimethylol propane was blended with the alumina (aluminum 2O3) 100 section as a cross linking agent per the solid-content (acrylic polymer) 100 section and as a thermally conductive filler, stirring mixture was improved, and the thermally conductive pressure sensitive adhesive was prepared. However, the viscosity elevation under manufacture or after manufacture is remarkable, and it \*\* at stability, and is \*\*. Although the adhesion sheet was produced like the example 1 using this thermally conductive pressure sensitive adhesive, a uniform sheet is obtained, and it is inside \*\*\*\*.

[0041] About each adhesion sheet produced in the above examples 1-6, it is \*\*\*\*\* by the following method about an adhesion test and a thermal resistance test. This result was made into Table 1.

[0042] The polyester film whose thickness is 25 micrometers was stuck on one side of a <adhesion-test> adhesion sheet, and the examination tape with a width of face [ of 20mm ] and a length of 100mm was produced. After having made 1 \*\*\*\* the stainless steel board which had this ground with 2kg roller, sticking and aging for 30 minutes on 23 degrees C and the conditions of 65%

of relative humidity, it pulled on 23 degrees C and the conditions of 65% of relative humidity, the testing machine tore off 180 degrees at 300mm hauling speed for /, and adhesive strength was measured.

[0043] The transistor in <thermal resistance test> TO-220 package is dipped in water using an adhesion sheet, and it is 2 the sticking-by-pressure pressure of 1kg/cm to a \*\*\*\*\* heat sink in constant temperature. After carrying out adhesion fixation, the output of a constant rate was supplied to the transistor and the temperature gradient (T2-T1) of the temperature (T2) of a transistor and the skin temperature (T1) of the adhesion sheet bottom was measured. From this temperature gradient, although it was made the following formula, an intermediary and thermal resistance were measured.

Thermal-resistance (cm<sup>2</sup> [ degree C and J/W) =(T2-T1) xA/PA: Area of a transistor (cm<sup>2</sup>)

P: Power consumption of a transistor (W)

[0044] In addition, the temperature (T2) of a transistor was measured with the thermocouple by which spot welding was carried out to the metal base portion of a transistor package. Moreover, the skin temperature (T1) of the adhesion sheet bottom made the minute hole in the heat sink, and measured it by pushing in a thermocouple. At that time, it approaches as much as possible and the thermocouple was held so that there might be no influence in the adhesion area of an adhesion sheet. In addition, the thermal resistance measured by the above-mentioned method means excelling in thermal conductivity again, so that the value is small.

[0045]

表 1

	接着力 (Kg/20mm幅)	熱抵抗 (℃・cm <sup>2</sup> /W)
実施例 1	1. 1	5. 0
実施例 2	1. 3	5. 2
実施例 3	1. 1	5. 0
実施例 4	1. 2	5. 5
実施例 5	1. 1	5. 3
実施例 6	1. 2	5. 6

[0046] The result of the above-mentioned table 1 shows that each of each adhesion sheets of the examples 1-6 of this invention has the outstanding adhesive property and thermal conductivity.

[0047]

[Effect of the Invention] As mentioned above, in this invention, since polymer with the polycarbonate structure used as adhesive polymer shows good dispersibility to various thermally conductive fillers with sufficient adhesive property, the thermally conductive pressure sensitive adhesive which does not receive any limit in the kind or loadings of a thermally conductive filler, therefore is excellent in both an adhesive property and thermal conductivity, and its adhesion sheets can be obtained easily.

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[Translation done.]